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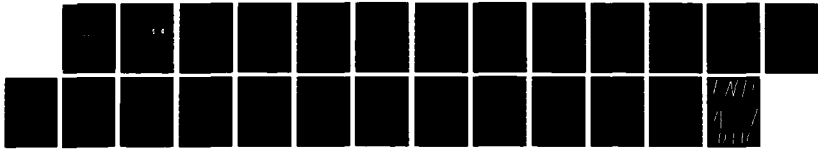

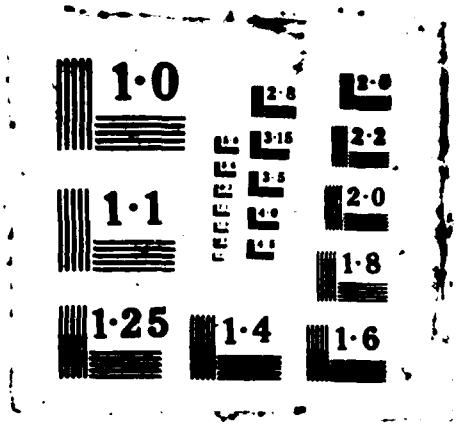
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The Plasmon Dispersion Relation on a Rough Surface: A Simple Approximation

by

Daniel Jelski and Thomas F. George

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This paper is concerned with periodic, laser-induced, chemical vapor deposition recently observed experimentally. In order to inquire further into this phenomena, it is first necessary to find a simple means of calculating the plasmon field strength for relatively deep gratings. The Rayleigh hypothesis is assumed, and only p-polarized, normally incident light is considered. A closed-form equation for the plasmon field intensity is then derived. Also discussed is the behavior of the plasmon dispersion relation for a shallow grating, but for a complex dielectric constant where the imaginary part is not necessarily small.

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THE PLASMON DISPERSION RELATION ON A ROUGH SURFACE: A SIMPLE APPROXIMATION

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ABSTRACT

This paper is concerned with periodic, laser-induced, chemical vapor deposition recently observed experimentally. In order to inquire further into this phenomena, it is first necessary to find a simple means of calculating the plasmon field strength for relatively deep gratings. The Rayleigh hypothesis is assumed, and only p-polarized, normally incident light is considered. A closed-form equation for the plasmon field intensity is then derived. Also discussed is the behavior of the plasmon dispersion relation for a shallow grating, but for a complex dielectric constant where the imaginary part is not necessarily small.

1. Introduction

Our laboratory has been investigating laser-induced chemical vapor deposition processes as recently observed experimentally.[1-3] In particular, we have been interested in the periodic structures observed by Brueck and Ehrlich[3]. In a later paper, we shall discuss this experiment directly. This paper, however, will be devoted to developing the theoretical framework necessary for the calculations.

The periodic structure that is observed evolves from the plasmon field induced by the laser. The laser interacts with the random surface roughness. Because the plasmon is resonant at one frequency (for shallow gratings), that frequency component grows as

$$\frac{\partial \xi}{\partial t} = aI(x), \tag{1}$$

where ξ is the grating amplitude, $I(x)$ is the total field intensity as a function of position, and a is the proportionality constant. From this it is clear that we need to develop expressions for the plasmon field intensity as a function of grating height and wavenumber. We can simplify the problem considerably in all that follows by assuming perpendicular incidence, in which case the x-dependence of the intensity depends only on the plasmon field and not on the reflected or incident fields. We can simplify the problem even further by using the Rayleigh expansion. This is an exact solution above the selvedge region and an approximation within the selvedge region. However, we are primarily interested in field strengths above the selvedge region, so that this expansion is justified.

Many authors have discussed the interaction of radiation with a rough surface or grating. Petit[4] has written a good introduction to the subject.

Maradudin[5] has discussed the behavior of surface polaritons and plasmons (which together are what we are generically referring to as a surface plasmon). In particular, he has worked out the theory for randomly rough surfaces and has written expressions for the dispersion relation using both the Rayleigh expansion and the exact formula using Green's functions. We shall make much use of this work as we go along. Other authors have also made significant contributions. Jha, Kirtley and Tsang[6] have developed the theory for a shallow grating. These authors have made extensive use of the work of Toigo, Marvin, Celli and Hill,[7,8] who have written the Rayleigh expansion in a remarkably simple form. Finally, Agassi and George[9,10] have developed a dressed Rayleigh expansion which eliminates many of the numerical difficulties encountered previously.

We are interested in the plasmon field intensity above the selvedge region for reasonably deep gratings. We need an expression as a function of both grating height and grating wavenumber. Since we shall also need the derivatives of this function, it is important to derive an expression in closed form. To date, all calculations for deep gratings have involved the inversion of large matrices. Given the conditions of our application, this is impractical. Therefore, we must develop an approximation which is still valid in the deep grating case, but which provides a closed form equation for the plasmon field strength. Two recent papers[11,12] have made progress in this field. In particular, the work by Weber[12] represents a generalization of our present result. We will compare our result with his in the conclusion.

We should also point out that there has been considerable work done on a closely related problem. Fauchet and Siegman[13] have discovered ripples as a result of laser annealing, and Sipe et al.[14] have developed an elegant

theory to account for this phenomena. But these are effects of surface damage rather than surface deposition, and hence the theoretical requirements are more stringent in that the Rayleigh hypothesis can no longer be applied.

But there is one additional problem. The experiment of Brueck and Ehrlich involved a plasmon wave along a cadmium surface. The dielectric constant for cadmium (at UV frequencies) is approximately

$$\epsilon = -2.5 + 1.3i. \tag{2}$$

Most calculations, even for shallow gratings, have assumed that the imaginary part of the dielectric constant is either zero[4,5], or small.[6] Since this is not the case for us, we shall digress a bit and discuss the effect on the dispersion relation as the imaginary part of the dielectric constant becomes large.

The remainder of this paper is organized as follows. In section 2 we shall discuss the dispersion relation as a function of dielectric constant. In section 3 we shall derive our approximate formula for the plasmon field strength. Section 4 contains a discussion of our results, including a numerical comparison of our approximation with the exact calculation.

2. Dispersion Relation

We can write the solution of the homogenous Helmholtz equation as

$$\underline{E}_a = E_i \underline{p}_{\ell-} \exp(i[kx-\alpha_0 z]) + \sum_{\ell=-\infty}^{\infty} A_\ell \underline{p}_{\ell+} \exp(i[k_\ell x+\alpha_\ell z]) \tag{3}$$

$$z>\xi(x)$$

$$\underline{E}_b = \sqrt{\epsilon}\,\underline{s} \sum_{\ell} C_\ell \exp(i[k_\ell x - \beta_\ell z]) \qquad (4)$$

$z<\xi(x)$,

where

$$k = \frac{\omega}{c} \qquad \epsilon = \epsilon_1 + i\epsilon_2 \qquad \epsilon_1 < -1$$

$$k_\ell = k_t + \ell k_g \qquad \alpha_\ell = (k^2 - k_\ell^2)^{1/2} \quad \begin{matrix} \mathrm{Im}\alpha > 0 \\ \alpha_0 > 0 \end{matrix} \qquad (5)$$

$$\beta_\ell = (\epsilon k^2 - k_\ell^2)^{1/2} \qquad \mathrm{Im}\beta_\ell > 0 \qquad \underline{p}_{\ell\pm} = \frac{1}{k}[k_\ell \underline{z} \mp \alpha_\ell \underline{x}]$$

and where k_g is the plasmon frequency and E_i the incident field strength. The notation has been culled from refs. 5 and 9. This solution is exact as long as one is outside the selvedge region. Within the selvedge region it may be a good approximation, but as mentioned previously, we are primarily concerned with the region outside, and hence we can use eqs. 4 and 5 without reservation. From the above, it is apparent that A_0 is the reflected field, while A_ℓ is the ℓ-th order Bragg reflection (α_ℓ real), or surface plasmon (α_1 imaginary). We are interested in the plasmon effect, and hence we shall always assume that $k_g > k$.

Now, suppose that the surface profile is sinusoidal and can be written as

$$\xi(x) = \xi\cos(k_g x) \,, \qquad (6)$$

and that ξ is small. Then, for perpendicularly incident, p-polarized light, we can write the the plasmon field strength as[6]

$$A_\ell = \frac{\alpha_0 \beta_\ell [k^2 \xi (\varepsilon \alpha_\ell - \beta_\ell)]}{\varepsilon k^2 - k_g^2 (1+\varepsilon)} \quad . \tag{7}$$

For small ξ, A_ℓ is very small unless the denominator is very small. If ε is real, the denominator, which we shall call the resonance factor and denote by R, is zero if

$$k_g^2 = k^2 \frac{\varepsilon_1}{\varepsilon_1 + 1} \quad . \tag{8}$$

This is the familiar plasmon resonance condition for a flat surface and a real dielectric constant. However, if ε is complex then the strength of the plasmon field depends on the modulus of R, and it is no longer sufficient merely to minimize the real part. We must then minimize

$$|R|^2 = [k_g^2(1+\varepsilon_1) - k^2\varepsilon_1]^2 + [\varepsilon_2(k_g^2 - k^2)]^2. \tag{9}$$

Differentiating with respect to k_g and setting the result to zero, we obtain a new dispersion relation

$$k_g^2 = \frac{k^2[\varepsilon_1(1+\varepsilon_1) - \varepsilon_2^2]}{(1+\varepsilon_1)^2 + \varepsilon_2^2}. \tag{10}$$

As $\epsilon_2 \to 0$ this reduces to eq. 5. We also note that if $\epsilon_2 \neq 0$, then $|R| \neq 0$, and the resonance is both dampened and broadened. Equation 10 is the flat surface plasmon dispersion relation for an arbitrary, complex-valued dielectric constant.

As the grating becomes deeper, R also depends on higher powers of ξ_1, whereby a branching occurs in the dispersion relation and the resonance becomes less pronounced. Thus the mechanism of the periodic deposition process should now be clear. In the beginning, the incident light is resonant with one frequency component of the randomly rough, but very shallow grating. Because of this resonance, the plasmon of that frequency is much stronger than the others. Hence the field intensity is periodic, and deposition, following eq. 1, has the same periodicity, leading to a growth in amplitude of the grating.

As the grating becomes larger, the dispersion relation branches, and the resonance is correspondingly broadened and dampened. This results in a decrease in the periodicity of the field intensity and a corresponding decrease in the differential rate of deposition (i.e., deposition occurs evenly, not necessarily more slowly). Hence the grating stops growing.

3. Calculation of Plasmon Field Intensity

We can write an expression for the coefficients of eq. 3 as[7,9]

$$\sum_{\ell=-\infty}^{\infty} M_{m,\ell} A_\ell = \mu_m E_i, \tag{11}$$

where

$$M_{m,\ell} = \frac{\alpha_m \beta_\ell + k_\ell k_m}{\alpha_\ell + \beta_m} (i)^{m-\ell} J_{m-\ell}(\xi[\alpha_\ell - \beta_m])\pi \qquad (12)$$

$$\mu_m = \frac{-\alpha_0 \beta_m + k_0 k_m}{\alpha_0 + \beta_m} (i)^m J(\xi[\alpha_0 + \beta_m]), \qquad (13)$$

and $J_m(x)$ is the m-th-order Bessel function. In general, this results in an infinite number of coupled equations, from which the A_ℓ's can be extracted. In practice, Agassi and George[9] have shown that 50 equations is sufficient to find an accurate solution. However, in our circumstance this is impossible. We must find the plasmon field strength as a function of both grating height and wavenumber, which means that we would have to invert two 50 x 50 real matrices for each data point. Using the approximation scheme which we shall derive, we have calculated 10,000 data points. This is perhaps more than necessary, but doing so has enabled us to thoroughly understand the behavior of these functions.

For our approximation, we borrow a page from Maradudin[5] and extend that result to higher orders. The method rests on two ideas. One is that if A_1 is a plasmon at resonance with k_g, then

$$A_1 >> A_m \qquad m \neq 1. \qquad (14)$$

The second condition is to note that

$$M_{nn} \sim 1, \quad M_{n,n\pm1} \sim \xi, \quad M_{n,n\pm2} \sim \xi^2, \ \ldots \qquad (15)$$

so that even for reasonably deep gratings we can assume that $M_{11} > M_{12} > M_{13}$ etc.

A special case of eq. 11 is

$$M_{11}A_1 + \sum_{m\neq 1} M_{1,m}A_m = \mu_1 E_i \tag{16}$$

or

$$A_1 = \frac{\mu_1 E_i + \sum_{m\neq 1} M_{1m}A_m}{M_{11}}. \tag{17}$$

Thus $R = M_{11}$, which must therefore go to zero if A_1 is to be resonant. Expanding A_{11} to first order in ξ yields our result of eq. 4.

Equation 17 is exact. Now we make our approximation. Suppose that $m \neq 1$. Then we can write

$$M_{mm}A_m + \sum_{p\neq m} M_{mp}A_p = \mu_m E_i \tag{18}$$

But because we are at resonance, of the sum over p, p=1 is by far the largest term. Thus we can simplify eq. 18 as

$$A_m = \frac{\mu_m E_i - M_{m,1}A_1}{M_{mm}}. \tag{19}$$

Substituting eq. 19 into 17, we obtain

$$A_1 = \frac{(\mu_1 - \sum_{p\neq 1} \frac{M_{1p}}{M_{pp}} \mu_p)E_i}{M_{11} - \sum_{p\neq 1} \frac{M_{1p}M_{p1}}{M_{pp}}}. \tag{20}$$

In the case of normal incidence, $A_1 = A_{-1}$ by symmetry, and we are not justified in leaving A_{-1} out of eq. 19. Inclusion of this term for normal incidence yields

$$A_1 = \frac{(\mu_1 - \sum_{p\neq 1} (M_{1p} + M_{1-p})\frac{\mu_p}{M_{pp}})E_i}{[(M_{11} + M_{1-1}) - \sum_{p\neq 1} (M_{1p} + M_{1-p})(M_{p1} + M_{p-1})\frac{1}{M_{pp}}} \qquad (21)$$

where p is summed only over non-negative integers. It is clear that in the shallow grating case, both eqs. 20 and 21 reduce to eq. 7.

We shall use eq. 21 with one modification: To ensure numerical convergence, we shall use the dressed Rayleigh expansion.[9] Thus we rewrite eq. 10 as

$$\sum_{\ell=-\infty}^{\infty} M^D_{m,\ell} A^D_\ell = \mu^D_m E_i \qquad (22)$$

where

$$M^D_{m,n} = M_{m,n}\exp(i\xi[\alpha_n + \beta_m]) \qquad (23)$$

$$\mu^D_m = \mu_m \exp(i\beta_m \xi), \qquad A^D_n = A_n \exp(i\alpha_n \xi).$$

This eliminates numerical instabilities, not only in the exact calculation, but also for our approximation. In practice, for the grating heights of interest, we have found that summing p from 0 to 15 in eq. 21 provides sufficient accuracy.

4. Conclusion

Equation 21, modified if necessary by the transformation of eq. 23, constitutes our closed form approximation for the plasmon field strength. Let us consider the advantages and shortcomings of our approximation. The first constraint is to notice that for our approximation to be valid, eq. 14 must hold. This means that we must be on the resonance frequency. Were we not on the resonance frequency, then the approximation used in deriving eq. 19 would not be valid. Secondly, our approximation still is a function of grating height. Note that we have used eq. 15, which means that we are discarding higher orders of ξ for non-resonant plasmons. As these become large, then our approximation becomes less accurate.

Nevertheless, we are happy to report that this method preserves the qualitative features of the full calculation. Figure 1 illustrates this very clearly. Here we have calculated the plasmon field strength ($|A_1|$) for various grating heights using both our new method and the full calculation. It is clear that for gratings up to $\xi \approx 25$ nm, the simple calculation preserves the qualitative features of the exact method. This corresponds to a value of $k_g \xi \approx .7$, which is a very deep grating. The quantitative picture may also be better than appears in the graph. The calculation was done for $k_g = 2.95 \times 10^7\ m^{-1}$. This is the resonance frequency for shallow gratings, but it is off resonance for deeper gratings. At 25 nm, we have observed that the resonance frequency is closer to $3.1 \times 10^{-7}\ m^{-1}$. We have ignored this shift in frequency in our calculations.

As mentioned previously, our result compares with the more general result of Weber.[12] His is more general in that it explicitly accounts for three terms in the Rayleigh expansion rather than just one. However, at resonance, the other two terms can be expected to contribute negligibly, and

hence our result should be nearly as accurate. Off resonance, of course, the other terms are significant and Weber's method is clearly preferable. Strict numerical comparison is impossible since he did his calculation using silver, with a small ϵ_2, whereas ours is for cadmium. Furthermore, his example is for a sawtooth grating while ours is for a sinusoidal grating. But it is very clear from a comparision of figure 1 with his table I that the same qualitative features hold, namely that the plasmon resonance intensity is underestimated. Our formalism, while not as general, is derived much more simply and is easier to use, and is probably just as accurate where applicable.

In a later paper[15] we shall apply this formalism to the problem of periodic, laser-induced, chemical vapor deposition. We should be able to predict the growth rate and maximum peak height using the above method.

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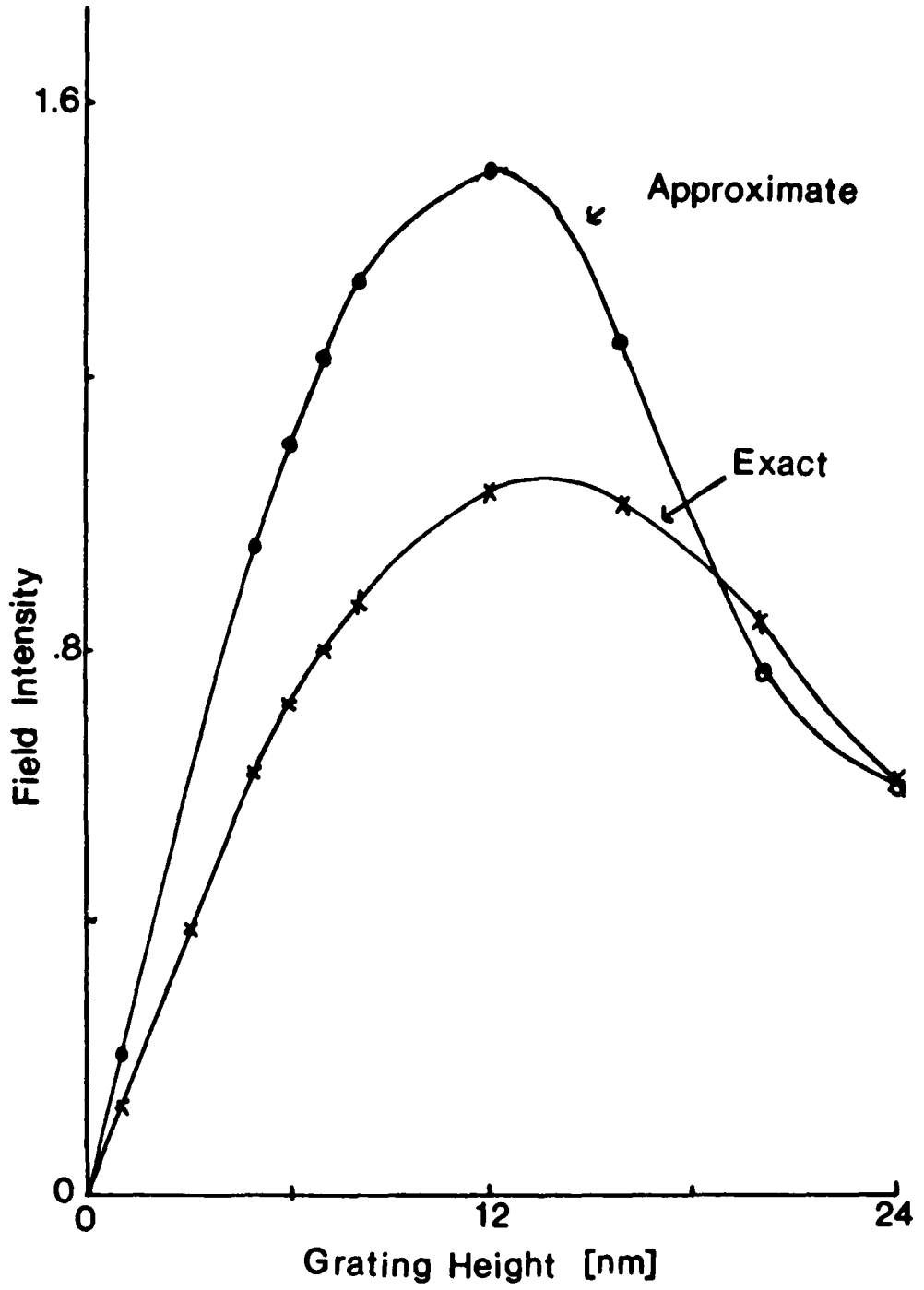
FIGURE CAPTION

Figure 1. This graph compares the plasmon field strength ($|A_1|$) calculated from eq. 21 (using the dressed Rayleigh expansion) with that from the exact method. The dielectric constant used is -2.5 + 1.3i, the incident light has a wavelength of 257 nm ($k = 2.44 \times 10^7\ m^{-1}$), and the grating wavenumber is taken as $2.95 \times 10^7\ m^{-1}$.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Field Intensity
Approximate
Exact
0
12
24
Grating Height [nm]

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